

論文内容の要旨

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In order to improve the photocatalytic activity of titanium dioxide (TiO_2), the use of non-metal dopants have been carried out in the last years. The incomplete oxidation of the Titanium Nitride (TiN) could result in a facile route towards N- TiO_2 . However, the fcc-structure of TiN_{1-X} can extend from 33 to 55 mol% of N.

The first step of this work lies in the development of a new method for the preparation of high dense TiN_{1-X} using pulsed electric current sintering and its mechanical characterization. After this, the oxidation of these TiN_{1-X} bulks was carried out by Air, O_2 -HIP and H_2O_2 . Finally, in order to improve the catalytic activity, the best oxidation processes were selected and applied to TiN_{1-X} powders.

Results in this work suggest that the dehydrogenation of TiH_2 and subsequent reaction with TiN by pulsed electric current sintering can be proposed as a good alternative for the preparation of high dense TiN_{1-X} bulks. The grain size of the bulks increased as the value of X in TiN_{1-X} also increased. Although the higher grain size, samples with high values of X in TiN_{1-X} showed the hardest values following different behavior from Hall-Petch relation and indicating the improvement of TiH_2 in both densification and hardness without high degradation in the fracture toughness.

Regarding with the oxidation results different oxidation conditions resulted in different N species in the TiO_2 structure. The oxidation of TiN_{1-X} bulks by H_2O_2 , which showed the best photocatalytic activities, resulted in the formation of anatase oxide ridges in the grain boundaries of the substrate. Two different types of substitutional N were observed by XPS which correspond with the N as dopant in anatase and the N in TiN_{1-X} . The discoloration of Methylene blue (MB) during photocatalysis decreased as X in TiN_{1-X} increased due to the recombination process produced by the existences of Ti^{3+} in these materials. The formation of anatase in the boundaries was promoted by the similarity in the crystalline structure with TiN. On the other hand, the transformation to rutile was also studied during the oxidation of TiN_{1-X} in air and O_2 -HIP. The formation to rutile was promoted at high X in TiN_{1-X} and at high pressures like in O_2 -HIP due to the existence of V_O produced by the formation of an oxynitride. The grown to rutile was observed through the plane (101) in both cases and it is considered to be due to the similar arrangement of the Ti sublattice in {100} of TiN_{1-X} . These results showed a good agreement with the photocatalytic activities in the lower discoloration rates of MB of those samples oxidized by O_2 -HIP with high transformation to rutile. Finally, the oxidation by H_2O_2 of TiN_{1-X} powders and subsequent thermal treatment at 450°C for 2 h resulted in the complete crystallization to anatase. However, the oxynitride produced by the oxidation of powders by H_2O_2 without thermal treatment showed complete discoloration of MB even at shorter periods of time.